

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
 REQUEST FOR FILING NATIONAL PHASE OF
PCT APPLICATION UNDER 35 U.S.C. 371 AND 37 CFR 1.494 OR 1.495

10/088307

To: Hon. Commissioner of Patents
 Washington, D.C. 20231



00909

TRANSMITTAL LETTER TO THE UNITED STATES
 DESIGNATED/ELECTED OFFICE (DO/EO/US)

Atty Dkt: P 0290749 /60381/UST
 M# /Client Ref.

From: Pillsbury Winthrop LLP, IP Group:

Date: March 18, 2002

This is a **REQUEST** for **FILING** a PCT/USA National Phase Application based on:

- | | | |
|------------------------------|------------------------------|-------------------------------------|
| 1. International Application | 2. International Filing Date | 3. Earliest Priority Date Claimed |
| <u>PCT/GB00/03113</u> | <u>14 August 2000</u> | <u>16 September 1999</u> |
| <u>↑ country code</u> | Day MONTH Year | Day MONTH Year |
| | | (use item 2 if no earlier priority) |
4. Measured from the earliest priority date in item 3, this PCT/USA National Phase Application Request is being filed within:
- (a) ☐ 20 months from above item 3 date (b) ☒ 30 months from above item 3 date,
- (c) Therefore, the due date (unextendable) is March 16, 2002
5. Title of Invention DISAZO DYES AND INKS CONTAINING THEM
6. Inventor(s) Mark Robert James

Applicant herewith submits the following under 35 U.S.C. 371 to effect filing:

7. ☒ Please immediately start national examination procedures (35 U.S.C. 371 (f)).
8. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2)) is transmitted herewith (file if in English but, if in foreign language, file only if not transmitted to PTO by the International Bureau) including:
- a. ☒ Request;
- b. ☒ Abstract;
- c. 16 pgs. Spec. and Claims;
- d. sheet(s) Drawing which are ☐ informal ☐ formal of size ☐ A4 ☐ 11"
9. ☒ A copy of the International Application has been transmitted by the International Bureau.
10. A translation of the International Application into English (35 U.S.C. 371(c)(2))
- a. ☐ is transmitted herewith including: (1) ☐ Request; (2) ☐ Abstract;
- (3) pgs. Spec. and Claims;
- (4) sheet(s) Drawing which are:
- ☐ informal ☐ formal of size ☐ A4 ☐ 11"
- b. ☒ is not required, as the application was filed in English.
- c. ☐ is not herewith, but will be filed when required by the forthcoming PTO Missing Requirements Notice per Rule 494(c) if box 4(a) is X'd or Rule 495(c) if box 4(b) is X'd.
- d. ☐ Translation verification attached (not required now).

RE: USA National Phase Filing of PCT /GB00/03113

11. ☒ Please see the attached Preliminary Amendment
12. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)), i.e., before 18th month from first priority date above in item 3, are transmitted herewith (file only if in English) including:
13. ☒ PCT Article 19 claim amendments (if any) have been transmitted by the International Bureau
14. ☐ Translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)), i.e., of **claim amendments** made before 18th month, is attached (required by 20th month from the date in item 3 if box 4(a) above is X'd, or 30th month if box 4(b) is X'd, or else amendments will be considered canceled).

15. **A declaration of the inventor** (35 U.S.C. 371(c)(4))
- a. ☒ is submitted herewith ☒ Original ☐ Facsimile/Copy
- b. ☐ is not herewith, but will be filed when required by the forthcoming PTO Missing Requirements Notice per Rule 494(c) if box 4(a) is X'd or Rule 495(c) if box 4(b) is X'd.

16. **An International Search Report (ISR):**

- a. Was prepared by ☒ European Patent Office ☐ Japanese Patent Office ☐ Other
- b. ☒ has been transmitted by the international Bureau to PTO.
- c. ☒ copy herewith (5 pg(s).) ☒ plus Annex of family members (1 pg(s).).

International Preliminary Examination Report (IPER):

- a. ☒ has been transmitted (if this letter is filed after 28 months from date in item 3) in English by the International Bureau with Annexes (if any) in original language.
- b. ☒ copy herewith in English.
- c.1 ☐ IPER Annex(es) in original language ("Annexes" are amendments made to claims/spec/drawings during Examination) including attached amended:
- c.2 ☐ Specification/claim pages # ___ claims # ___ Dwg Sheets # ___
- d. ☐ Translation of Annex(es) to IPER (required by 30th month due date, or else annexed amendments will be considered canceled).

Information Disclosure Statement including:

- a. ☒ Attached Form PTO-1449 listing documents
- b. ☒ Attached copies of documents listed on Form PTO-1449
- c. ☒ A concise explanation of relevance of ISR references is given in the ISR.

19. ☒ **Assignment** document and Cover Sheet for recording are attached. Please mail the recorded assignment document back to the person whose signature, name and address appear at the end of this letter.
20. ☐ Copy of Power to IA agent.
21. ☐ **Drawings** (complete only if 8d or 10a(4) not completed): ___ sheet(s) per set: ☐ 1 set informal; ☐ Formal of size ☐ A4 ☐ 11"

22. Small Entity Status ☒ is **Not** claimed ☐ is claimed (pre-filing confirmation required)
- 22(a) ___ (No.) Small Entity Statement(s) enclosed (since 9/8/00 Small Entity Statements(s) not essential)

23. **Priority** is hereby claimed under 35 U.S.C. 119/365 based on the priority claim and the certified copy, both filed in the International Application during the international stage based on the filing in (country) Great Britain of:

	<u>Application No.</u>	<u>Filing Date</u>		<u>Application No.</u>	<u>Filing Date</u>
(1)	9921928.9	16 Sept 1999	(2)		
(3)			(4)		
(5)			(6)		

- a. ☒ See Form PCT/IB/304 sent to US/DO with copy of priority documents. If copy has not been received, please proceed promptly to obtain same from the IB.
- b. ☐ Copy of Form PCT/IB/304 attached.

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JC10 Rec'd PCT/PTO 1 8 MAR 2002

RE: USA National Phase Filing of PCT/GB00/03113

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24. Attached: Preliminary Amendment

25. Per Item 17.c2, **cancel original** pages #__, claims #__, Drawing Sheets #26. **Calculation of the U.S. National Fee (35 U.S.C. 371 (c)(1)) and other fees is as follows:**Based on amended claim(s) per above item(s) ☐ 12, ☐ 14, ☐ 17, ☐ 25 (hilitte)

Total Effective Claims	20	minus 20 =	0	x \$18/\$9	=	\$0	966/967
Independent Claims	1	minus 3 =	0	x \$84/\$42	=	\$0	964/965
If any proper (ignore improper) Multiple Dependent claim is present,				add \$280/\$140	+0		968/969

BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(4)): →→ **BASIC FEE REQUIRED, NOW** →→→→A. If country code letters in item 1 are **not** "US", "BR", "BB", "TT", "MX", "IL", "NZ", "IN", "ZA", "LC" or "PH"

See item 16 re:

1. Search Report was <u>not</u> prepared by EPO or JPO -----	add \$1,040/\$520		960/961
2. Search Report was prepared by EPO or JPO -----	add \$890/\$445	+890	970/971

SKIP B, C, D AND E UNLESS country code letters in item 1 are "US", "BR", "BB", "TT", "MX", "IL", "NZ", "IN", "ZA", "LC" or "PH"

→ ☐ B. If USPTO did not issue both International Search Report (ISR) and (if box 4(b) above is X'd) the International Examination Report (IPER), ----- add \$1,040/\$520 +0 960/961

→ ☐ C. If USPTO issued ISR but not IPER (or box 4(a) above is X'd), ----- add \$740/\$370 +0 958/959

→ ☐ D. If USPTO issued IPER but IPER Sec. V boxes not all 3 YES, ----- add \$710/\$355 +0 956/957

→ ☐ E. If international preliminary examination fee was paid to USPTO and Rules 492(a)(4) and 496(b) satisfied (i.e., in IPER Sec. V all 3 boxes must be YES for all claims), -- add \$100/\$50 +0 962/963

27. **SUBTOTAL =** \$89028. If Assignment box 19 above is X'd, add Assignment Recording fee of ----\$40 +40 (581)29. If box 15a is x'd, determine whether inventorship on Declaration is different than in international stage. If yes, add (per Rule 497(d)) ----\$130 +0 (098)30. Attached is a check to cover the ----- **TOTAL FEES** \$930

Our Deposit Account No. 03-3975

Our Order No. 070662 | 0290749
C# M#

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CHARGE STATEMENT: The Commissioner is hereby authorized to charge any fee specifically authorized hereafter, or any missing or insufficient fee(s) filed, or asserted to be filed, or which should have been filed herewith or concerning any paper filed hereafter, and which may be required under Rules 16-18 and 492 (missing or insufficient fee only) now or hereafter relative to this application and the resulting Official document under Rule 20, or credit any overpayment, to our Account/Order Nos. shown above for which purpose a duplicate copy of this sheet is attached.

This CHARGE STATEMENT does not authorize charge of the issue fee until/unless an issue fee transmittal form is filedPillsbury Winthrop LLP
Intellectual Property GroupBy Atty: Richard A. Steinberg Reg. No. 26,588Sig: Richard A. Steinberg Fax: (703) 905-2500
Tel: (703) 905-2039

Atty/Sec: RAS/kmh

10/088307

JC10 Rec'd PCT/PTO 1 8 MAR 2002

#3/a

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re PATENT APPLICATION OF

BOX PCT

Inventor: Mark Robert James

Filed: March 18, 2002

Title: *DISAZO DYES AND INKS CONTAINING THEM*

PRELIMINARY AMENDMENT

Hon. Commissioner of Patents
Washington, D.C. 20231

Sir:

Prior to prosecution on the merits, please amend this application as follows herein.

In the Title

Please delete the existing title and insert the following new title:
--DISAZO DYES AND INKS CONTAINING THEM--.

In the Specification

Please insert the following heading and paragraph after the title of the application on page 1 of the specification:

--Cross Reference to Related Applications

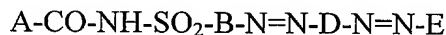
This application is a national phase application based on PCT/GB00/03113, filed August 14, 2000, and which further claims priority from British Application No. 9921928.9 filed September 16, 1999. These applications in their entirety are incorporated herein by reference.--

10088307 "031802"

In the Claims

Kindly amend the claims as follows:

3. (Amended) A compound according to claim 1 of Formula (1) and salts thereof:



Formula (1)

wherein:

A is an optionally substituted alkyl or aryl group;

B and D are each independently optionally substituted phenylene or naphthylene;
and

E is optionally substituted naphthylene.

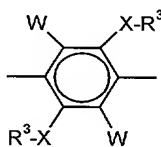
4. (Amended) A compound according to claim 1 wherein A is optionally substituted C₁₋₄-alkyl or optionally substituted phenyl.

5. (Amended) A compound according to claim 3 wherein B is optionally substituted phenylene.

6. (Amended) A compound according to claim 3 wherein B is optionally substituted 1,4- phenylene.

7. (Amended) A compound according to claim 3 wherein D is optionally substituted 1,4- phenylene.

8. (Amended) A compound according to claim 3 wherein D is of the formula:



wherein each:

W independently is H, C₁₋₄-alkyl, alkoxy, polyalkylene oxide, nitro, cyano, amino, sulpho, halo, ureido, SO₂F, PO₄H₂, hydroxy, carboxy, ester, -NR¹R², -COR¹, -CONR¹R² or -SO₂NR¹R², wherein R¹ and R² are each independently H or alkyl;

X independently is O, NH or S;

R³ independently is H or optionally substituted alkyl or optionally substituted aryl.

9. (Amended) A compound according to claim 3 wherein D is naphthylene carrying one or more groups selected from hydroxy, amino, sulpho and carboxy.

10. (Amended) A compound according to claim 3 wherein E is naphthylene carrying one or more substituents selected from hydroxy, amino, sulpho and carboxy.

11. (Amended) A compound according to claim 3 wherein E is a 1-hydroxy-3-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position; or a 1-hydroxy-5-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position; or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position

12. (Amended) A compound according to claim 1 wherein the pKa of the -NH-group is from 8 to 3.

13. (Amended) A composition comprising a compound according to claim 1 and a liquid medium.

16. (Amended) A paper, an overhead projector slide or a textile material printed with an ink according to claim 14.

18. (Amended) A method for the colouration of a substrate which comprises treating with a compound according to claim 1.

Please refer to the attached Appendix for changes made to the above claims.

100883307 "031802"

Please add the following new claims:

--19. (New) A paper, an overhead projector slide or a textile material printed with an ink of a compound according to claim 1.

20. (New) A paper, an overhead projector slide or a textile material printed by means of a process according to claim 15.--

10088307.031802

REMARKS

The present amendment adds reference to the priority applications to the specification and further amends Claims 3-13, 16 and 18 to remove multiple dependencies from the claims. No new matter has been added.

Favorable action on the merits is respectfully requested.

Respectfully submitted,

PILLSBURY WINTHROP LLP

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Date: March 18, 2002
Attorney Reference: 070662/0290749

Attachment: Appendix

10088307.031802

APPENDIX: VERSION TO SHOWS CHANGES MADE TO APPLICATION

In the Title

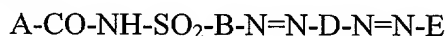
The existing title is deleted and the new title inserted as follows:

--DISAZO DYES AND INKS CONTAINING THEM--.

In the Claims

The claims were amended as follows:

3. (Amended) A compound according to [any one of the preceding claims] claim 1 of Formula (1) and salts thereof:



Formula (1)

wherein:

A is an optionally substituted alkyl or aryl group;

B and D are each independently optionally substituted phenylene or naphthylene;
and

E is optionally substituted naphthylene.

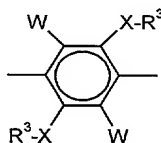
4. (Amended) A compound according to [any one of the preceding claims] claim 1 wherein A is optionally substituted C₁₋₄-alkyl or optionally substituted phenyl.

5. (Amended) A compound according to [any one of claim 3 or claim 4] claim 3 wherein B is optionally substituted phenylene.

6. (Amended) A compound according to [any one of claims 3 to 5] claim 3 wherein B is optionally substituted 1,4- phenylene.

7. (Amended) A compound according to [any one of claims 3 to 5] claim 3 wherein D is optionally substituted 1,4- phenylene.

8. (Amended) A compound according to [any one of claims 3 to 6] claim 3 wherein D is of the formula:



wherein each:

W independently is H, C₁₋₄-alkyl, alkoxy, polyalkylene oxide, nitro, cyano, amino, sulpho, halo, ureido, SO₂F, PO₄H₂, hydroxy, carboxy, ester, -NR¹R², -COR¹, -CONR¹R² or -SO₂NR¹R², wherein R¹ and R² are each independently H or alkyl;

X independently is O, NH or S;

R³ independently is H or optionally substituted alkyl or optionally substituted aryl.

9. (Amended) A compound according to [any one of claims 3 to 6] claim 3 wherein D is naphthylene carrying one or more groups selected from hydroxy, amino, sulpho and carboxy.

10. (Amended) A compound according to [any one of claims 3 to 9] claim 3 wherein E is naphthylene carrying one or more substituents selected from hydroxy, amino, sulpho and carboxy.

11. (Amended) A compound according to [any one of claims 3 to 11] claim 3 wherein E is a 1-hydroxy-3-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position; or a 1-hydroxy-5-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position; or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position

12. (Amended) A compound according to [any one of the preceding claims] claim 1 wherein the pKa of the -NH- group is from 8 to 3.

13. (Amended) A composition comprising a compound according to [any one of claims 1 to 12] claim 1 and a liquid medium.

16. (Amended) A paper, an overhead projector slide or a textile material printed with an ink according to claim 14[, with a compound according to any one of claims 1 to 12 or by means of a process according to claim 15].

18. (Amended) A method for the colouration of a substrate which comprises treating with a compound according to [any one of claims 1 to 12] claim 1.

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COMPOUND

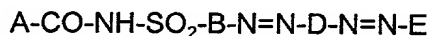
This invention relates to compoundss, to inks comprising these compounds, to ink jet printer cartridges containing these inks and to their use in ink jet printing ("IJP").

IJP is a non-impact printing technique in which droplets of ink are ejected through a fine nozzle onto a substrate without bringing the nozzle into contact with the substrate.

There are many demanding performance requirements for dyes and inks used in IJP. For example they desirably provide sharp, non-feathered images having good water-fastness, light-fastness and optical density. The inks are often required to dry quickly when applied to a substrate to prevent smudging, but they should not form a crust over the tip of an ink jet nozzle because this will stop the printer from working. The inks should also be stable to storage over time without decomposing or forming a precipitate which could block the fine nozzle.

According to the present invention there is provided a compound comprising a disazo chromophore and an optionally substituted acyl group linked together through a -SO₂NH- group.

Preferably where the nitrogen atom of the -SO₂NH- group is attached to the acyl group through a single covalent bond. It is especially preferred that the compound is of Formula (1) and salts thereof:



Formula (1)

wherein:

A is an optionally substituted alkyl or aryl group;

B and D are each independently optionally substituted phenylene or naphthylene;
and

E is optionally substituted naphthylene.

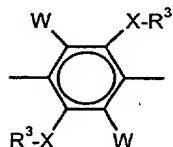
A is preferably an optionally substituted alkyl or aryl group it is especially preferred that A is optionally substituted C₁₋₄-alkyl or optionally substituted phenyl. Preferred optional substituents are selected from; alkoxy, more preferably C₁₋₄-alkoxy; polyalkylene oxide; nitro; cyano; amino; sulpho; halo, especially bromo, chloro or fluoro; ureido; PO₄H₂ SO₂F; hydroxy; carboxy; ester, more preferably -CO₂(C₁₋₄-alkyl); and -NR¹R²-COR¹, -CONR¹R² and -SO₂NR¹R² wherein R¹ and R² are each independently H or alkyl, preferably H or C₁₋₄-alkyl.

Preferably B is optionally substituted phenylene, more preferably optionally substituted 1,4-phenylene.

Preferred optional substituents which may be present on B, D or E are selected from alkyl, more preferably C₁₋₄-alkyl; alkoxy, more preferably C₁₋₄-alkoxy; polyalkylene oxide; nitro; cyano; amino; sulpho; halo, especially bromo, chloro or fluoro; ureido; PO₄H₂;

SO₂F; hydroxy; carboxy; ester, more preferably -CO₂(C₁₋₄-alkyl); and -NR¹R²-COR¹, -CONR¹R² and -SO₂NR¹R² wherein R¹ and R² are each independently H or alkyl, preferably H or C₁₋₄-alkyl.

Preferably D is an optionally substituted 1,4-phenylene group, more preferably D is of the formula:



wherein each:

W independently is H, optionally substituted alkyl, optionally substituted alkoxy, polyalkylene oxide, nitro, cyano, sulpho, halo, ureido, SO₂F, PO₄H₂, hydroxy, carboxy, ester, -NR¹R², -COR¹, -CONR¹R² or -SO₂NR¹R²;

X independently is O, NH or S;

R¹ and R² are each independently H or alkyl, preferably H or C₁₋₄-alkyl; and

R³ independently is H, optionally substituted alkyl or optionally substituted aryl.

Preferably each W independently is H, C₁₋₄-alkyl or C₁₋₄-alkoxy more preferably H, methyl, ethyl, methoxy or ethoxy, especially H. Preferably both groups represented by W are H and X is O.

Preferably each R³ independently is optionally substituted C₁₋₄-alkyl or optionally substituted phenyl more preferably C₁₋₄-alkyl or C₁₋₄-alkyl-OH, phenyl or phenyl carrying one or two substituents selected from carboxy and sulpho.

When D is also optionally substituted naphthylene it preferably carries one or more (preferably 1 to 4) substituents selected from hydroxy, amino, sulpho and carboxy.

Preferably E is optionally substituted naphthylene, more preferably carrying one or more (preferably from 1 to 4) groups selected from hydroxy, amino, sulpho and carboxy. More preferably E is naphthylene carrying one hydroxy group, one amino group and 0, 1 or 2 sulpho groups. It is especially preferred that E is a 1-hydroxy-3-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position; or a 1-hydroxy-5-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position; or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position.

Preferred optionally substituted amino groups are of the formula -NR⁴R⁵ where R⁴ and R⁵ are independently selected from H, optionally substituted alkyl and optionally substituted phenyl. Preferably R⁴ and R⁵ are both H.

The compounds described hereinbefore are preferably soluble in water. To achieve this solubility, the compounds preferably have 1, 2 or 3 water-dispersing

substituents. Preferred water-dispersing substituents are polyalkylene oxides and more preferably sulpho and carboxy.

Preferred polyalkylene oxide groups which may be present on A, B, D and/or E are poly(C_{2-3} -alkylene oxide) groups, more preferably polyethylene glycol or polypropylene glycol, preferably having from 1 to 20 glycol units and terminated with a hydroxy or C_{1-4} -alkoxy group.

Preferably A, B, D and E are selected from the definitions given above such that the pKa of the linking -NH- group (e.g as shown in Figure 1) is from 8 to 3, more preferably 7.5 to 4, especially 7 to 4.5. These preferences arise from the finding that compounds having such an -NH- group have a particularly good combination of water-fastness and optical density when printed on paper.

Any acid or basic groups on the compound, particularly carboxylic acid and sulphonic acid groups, are preferably in the form of a salt. Thus the Formulae shown herein include the compounds in free acid and in salt form.

The compounds of the invention may be prepared by condensing a diazo chromophore and an optionally substituted acyl compound, where one has a sulphonyl halide substituent and the other has an amino substituent. This condensation is preferably performed in the presence of base to absorb hydrogen halide as it is generated during the condensation.

The compound of Formula $A-CO-NH-SO_2-B-N=N-D-N=N-E$ may be prepared by diazotising an amine of the formula $A-CO-NH-SO_2-B-N=N-D-NH_2$ and coupling the resultant diazonium salt with a compound of the formula H-E.

The compound of Formula $A-CO-NH-SO_2-B-N=N-D-NH_2$ may be prepared by diazotising an amine of the formula $A-CO-NH-SO_2-B-NH_2$ and coupling the resultant diazonium salt with an amine of the formula H-D-NH₂. In some cases it may be necessary for the amino group to carry a protecting group which should be removed on completion of the reaction.

Many compounds of the formula $A-CO-NH-SO_2-B-NH_2$ are commercially available.

Preferably they may be prepared by condensing an amine of formula $B-(NH_2)_2$, in which one of the amino groups is protected, with a compound of formula $Cl-SO_2-NH-CO-A$ followed by removal of the protecting group.

More preferably they may be prepared by condensing an amine of formula $A-CO-NH_2$ with a compound of formula $Cl-SO_2-B-NH_2$ carrying a protecting group on the amino group of the $Cl-SO_2-B-NH_2$ compound, followed by removal of the protecting group.

Diazotisation is preferably performed at a temperature below 6°C, more preferably at a temperature in the range -10°C to 5°C. Preferably diazotisation is performed in water, preferably at a pH below 7 for the monoazo intermediate and above pH7 to form compounds of Formula (1). Dilute mineral acid, e.g. HCl or H₂SO₄, may be used to achieve the desired acidic conditions.

In the above processes, A, B, D and E are as hereinbefore defined.

Any acid or basic groups on the compound, particularly carboxylic acid and sulphonic acid groups, are preferably in the form of a salt. Thus the Formulae shown herein include the compounds in free acid and in salt form.

5 Preferred salts are alkali metal salts, especially lithium, sodium and potassium salts, ammonium and substituted ammonium salts. Especially preferred salts are salts with ammonia and volatile amines. The compounds may be converted into a salt using known techniques. For example, an alkali metal salt of a compound may be converted
10 into a salt with ammonia or an amine by dissolving an alkali metal salt of the compound in water, acidifying with a mineral acid and adjusting the pH of the solution to pH 9 to 9.5 with ammonia or the amine and removing the alkali metal cations by dialysis or by treatment with an ion exchange resin.

The compounds may exist in tautomeric forms other than those shown in this specification. These tautomers are included within the scope of the present claims. The compounds are particularly useful as dyes, especially for ink jet printing compositions.

According to a second aspect of the present invention there is provided a composition comprising a compound according to the first aspect of the invention, preferably of Formula (1) and a liquid medium.

Preferred compositions comprise:

20 (a) from 0.01 to 30 parts of a compound according to the first aspect of the invention, preferably of Formula (1); and

(b) from 70 to 99.99 parts of a liquid medium;

wherein all parts are by weight and the number of parts of (a)+(b)=100.

25 The number of parts of component (a) is preferably from 0.1 to 20, more preferably from 0.5 to 15, and especially from 1 to 5 parts. The number of parts of component (b) is preferably from 99.9 to 80, more preferably from 99.5 to 85, especially from 99 to 95 parts.

30 Preferably component (a) is completely dissolved in component (b). Preferably component (a) has a solubility in component (b) at 20°C of at least 10%. This allows the preparation of liquid dye concentrates which may be used to prepare more dilute inks and reduces the chance of the dye precipitating if evaporation of the liquid medium occurs during storage.

Preferred liquid media include water, a mixture of water and an organic solvent and an organic solvent free from water.

35 When the medium comprises a mixture of water and an organic solvent, the weight ratio of water to organic solvent is preferably from 99:1 to 1:99, more preferably from 99:1 to 50:50 and especially from 95:5 to 80:20.

It is preferred that the organic solvent present in the mixture of water and organic solvent is a water-miscible organic solvent or a mixture of such solvents. Preferred water-miscible organic solvents include C₁₋₆-alkanols, preferably methanol, ethanol, n-propanol,

isopropanol, n-butanol, sec-butanol, tert-butanol, n-pentanol, cyclopentanol and cyclohexanol; linear amides, preferably dimethylformamide or dimethylacetamide; ketones and ketone-alcohols, preferably acetone, methyl ether ketone, cyclohexanone and diacetone alcohol; water-miscible ethers, preferably tetrahydrofuran and dioxane; diols, preferably diols having from 2 to 12 carbon atoms, for example pentane-1,5-diol, ethylene glycol, propylene glycol, butylene glycol, pentylene glycol, hexylene glycol and thiodiglycol and oligo- and poly-alkyleneglycols, preferably diethylene glycol, triethylene glycol, polyethylene glycol and polypropylene glycol; triols, preferably glycerol and 1,2,6-hexanetriol; mono-C₁₋₄-alkyl ethers of diols, preferably mono-C₁₋₄-alkyl ethers of diols having 2 to 12 carbon atoms, especially 2-methoxyethanol, 2-(2-methoxyethoxy)ethanol, 2-(2-ethoxyethoxy)-ethanol, 2-[2-(2-methoxyethoxy)ethoxy]ethanol, 2-[2-(2-ethoxyethoxy)-ethoxy]-ethanol and ethyleneglycol monoallylether; cyclic amides, preferably 2-pyrrolidone, N-methyl-2-pyrrolidone, N-ethyl-2-pyrrolidone, caprolactam and 1,3-dimethylimidazolidone; cyclic esters, preferably caprolactone; sulphoxides, preferably dimethyl sulphoxide and sulpholane. Preferably the liquid medium comprises water and 2 or more, especially from 2 to 8, water-soluble organic solvents.

Especially preferred water-soluble organic solvents are cyclic amides, especially 2-pyrrolidone, N-methyl-pyrrolidone and N-ethyl-pyrrolidone; diols, especially 1,5-pentane diol, ethyleneglycol, thiodiglycol, diethyleneglycol and triethyleneglycol; and mono- C₁₋₄-alkyl and C₁₋₄-alkyl ethers of diols, more preferably mono- C₁₋₄-alkyl ethers of diols having 2 to 12 carbon atoms, especially 2-methoxy-2-ethoxy-2-ethoxyethanol.

Although not usually necessary, further colorants may be added to the ink to modify the shade and performance properties. Examples of such colorants include C.I.Direct Yellow 86, 132, 142 and 173; C.I.Direct Blue 199, and 307; C.I.Food Black 2; C.I.Direct Black 168 and 195; C.I.Acid Yellow 23; and any of the dyes used in ink jet printers sold by Seiko Epson Corporation, Hewlett Packard Company, Canon Inc. & Lexmark International. Addition of such further dyes can increase overall solubility leading to less kogation (nozzle blockage) for the resultant ink.

Examples of further suitable liquid media comprising a mixture of water and one or more organic solvents are described in US 4,963,189, US 4,703,113, US 4,626,284 and EP 4,251,50A.

When the liquid medium comprises an organic solvent free from water, (i.e. less than 1% water by weight) the solvent preferably has a boiling point of from 30° to 200°C, more preferably of from 40° to 150°C, especially from 50 to 125°C. The organic solvent may be water-immiscible, water-miscible or a mixture of such solvents. Preferred water-miscible organic solvents are any of the hereinbefore described water-miscible organic solvents and mixtures thereof. Preferred water-immiscible solvents include, for example, aliphatic hydrocarbons; esters, preferably ethyl acetate; chlorinated hydrocarbons, preferably CH₂Cl₂; and ethers, preferably diethyl ether; and mixtures thereof.

When the liquid medium comprises a water-immiscible organic solvent, preferably a polar solvent is included because this enhances solubility of the compound in the liquid medium. Examples of polar solvents include C₁₋₄-alcohols. In view of the foregoing preferences it is especially preferred that where the liquid medium is an organic solvent free from water it comprises a ketone (especially methyl ethyl ketone) &/or an alcohol (especially a C₁₋₄-alkanol, more especially ethanol or propanol).

The organic solvent free from water may be a single organic solvent or a mixture of two or more organic solvents. It is preferred that when the medium is an organic solvent free from water it is a mixture of 2 to 5 different organic solvents. This allows a medium to be selected which gives good control over the drying characteristics and storage stability of the ink.

Liquid media comprising an organic solvent free from water are particularly useful where fast drying times are required and particularly when printing onto hydrophobic and non-absorbent substrates, for example plastics, metal and glass.

The liquid media may also contain additional components conventionally used in ink jet printing inks, for example viscosity and surface tension modifiers, corrosion inhibitors, biocides, kogation reducing additives and surfactants which may be ionic or non-ionic.

It is preferred that a composition according to the second aspect of the invention is an ink or liquid dye concentrate.

A third aspect of the invention provides a process for forming an image on a substrate comprising applying an ink according to the second aspect of the invention thereto by means of an ink jet printer.

The ink used in this process is preferably a composition as defined in the second aspect of the present invention.

The inks preferably have a total concentration of divalent metal ions and trivalent metal ions below 1000 more preferably below 100, especially below 20, more especially below 10 parts per million by weight relative to the total weight of ink. Pure inks of this type may be prepared by using high purity ingredients and/or by purifying the ink after it has been prepared. Suitable purification techniques are well known, e.g. ultrafiltration, reverse osmosis, ion exchange and combinations thereof.

The ink jet printer preferably applies the ink to the substrate in the form of droplets which are ejected through a small orifice onto the substrate. Preferred ink jet printers are piezoelectric ink jet printers and thermal ink jet printers. In thermal ink jet printers, programmed pulses of heat are applied to the ink in a reservoir by means of a resistor adjacent to the orifice, thereby causing the ink to be ejected in the form of small droplets directed towards the substrate during relative movement between the substrate and the orifice. In piezoelectric ink jet printers the oscillation of a small crystal causes ejection of the ink from the orifice.

The substrate is preferably paper, plastic, a textile, metal or glass, more preferably paper, an overhead projector slide or a textile material, especially paper.

Preferred papers are plain or treated papers which may have an acid, alkaline or neutral character.

5 A fourth aspect of the present invention provides a paper, an overhead projector slide or a textile material printed with a composition according to the second aspect of the invention, a compound according to the first aspect of the invention or by means of a process according to third aspect of the invention.

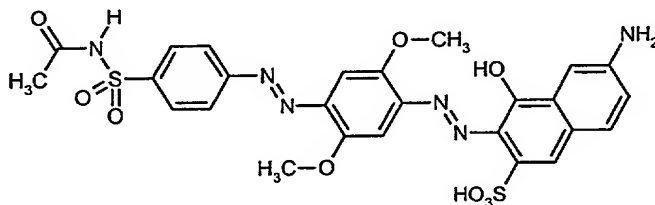
10 A fifth aspect of the present invention provides an ink jet printer cartridge comprising a chamber and an ink wherein the ink is in the chamber and the ink is according to the second aspect of the present invention.

A final aspect of the invention is a method for the colouration of a substrate which comprises treating with a compound according to the first aspect of this invention.

15 The invention is further illustrated by the following Examples in which all parts and percentages are by weight unless otherwise stated.

Example 1

Preparation of:



25 Sulfacetamide (16.8 parts) was dissolved in distilled water (550 parts) by the addition of the minimum amount of 47% caustic liquor with stirring. Sodium nitrite (5.68 parts) was then added and the solution was cooled to 0°C. This mixture was poured into pre-cooled (0°C) concentrated hydrochloric acid (75 parts) with vigorous stirring. The mixture was stirred at 0-5°C for 4 hours, then enough sulfamic acid was added to destroy the excess nitrous acid present and the mixture was stirred for a further 15 minutes. This mixture was then added to a stirred mixture of 2,5-dimethoxyaniline (12.6 parts) and methylated sprits (74OP, 400 parts) over approximately 15 minutes with the co-addition of enough ice and saturated sodium acetate solution to maintain the temperature at 0-5°C and the pH in the range of 4-5. The mixture was then stirred at 0-5°C for 1 hour, allowed to warm to room temperature and stirred over night at room temperature. The product was filtered, washed twice with distilled water and dried at approximately 70°C overnight to yield 28.5 parts of product at 91% strength by C, H and N analysis.

35

The monoazo intermediate (3 parts) and sodium nitrite (0.52 parts) was stirred in distilled water (100 parts) at 0°C. Concentrated hydrochloric acid (7.5 parts) was added with vigorous stirring and the mixture was stirred at 0-5°C for 2 hours. Tetrahydrofuran (150 parts) was added and the mixture was stirred at 0-5°C for a further 3 hours.

5 Sulfamic acid was added to destroy the excess nitrous acid present and the mixture was stirred for a further 15 minutes. This mixture was then added to a stirred solution of gamma acid (1.9 parts) and sodium carbonate (to pH 10-11) in water (100 parts), over approximately 30 minutes with the co-addition of enough ice and 47% caustic liquor solution to maintain the temperature at 0-5°C and the pH in the range of 9.5-10. The
10 mixture was stirred at 0-5°C for 1 hour, allowed to warm to room temperature and stirred overnight at room temperature. The mixture was acidified to pH 3 by the slow addition of concentrated hydrochloric acid, the product was filtered, washed with dilute hydrochloric acid and triturated twice with acetone (170 parts) containing 880 ammonia (3 drops).

The crude sodium salt was then exchanged for the ammonium salt by dissolving the compound in aqueous ammonia at approximately pH 10-11 and slowly pouring this solution into an equal volume of vigorously stirred 2N hydrochloric acid. The precipitated compound was filtered and the ion exchange process repeated.

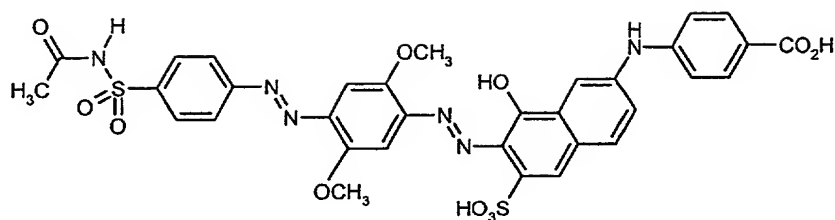
The crude ammonium salt was dissolved in dilute aqueous ammonia at approximately pH 10-11, filtered through a GF-A and GF-F glass fibre filter and dialysed using 'Visking tubing'. This solution was dried at approximately 70°C overnight to yield 1.8 parts of product which shown to have the correct structure by nmr and mass spectral analysis.

Inks were then prepared by dissolving the specified % of the compound in a stock solution of 9 parts water and 1 part 1-methyl-2-pyrrolidinone with the addition of
25 concentrated ammonia to give a pH of 9-10. The inks were then filtered through a 0.45 micron membrane filter and printed using HP 560 thermal IJ printer. The following results were obtained on Wiggins Conquer paper:

% Dye in ink	OD	Run down (24 h)
2.5	1.446	4

30 * OD means optional density of the printed paper, as measured by an x-rite spectrometer.

Rundown was measured by printing the ink in parallel bands onto the paper,
35 allowing the printed paper to dry for 24 hours, placing the printed paper at an angle of 45° and pouring 0.25ml of water down the paper. The run down (a measure of wet-fastness) was given a score of 1 to 10, where 10 means no visible ink run (high wet-fastness).

Example 2Preparation of:

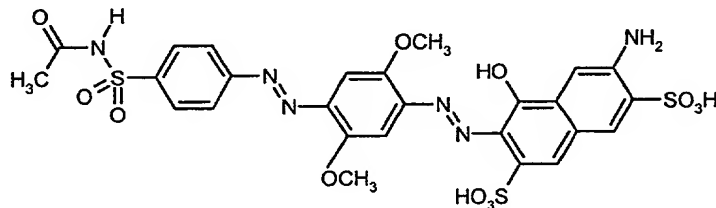
5

Example 1 was repeated except that p-carboxyphenyl gamma acid was used in place of gamma acid in the final coupling reaction.

Inks containing the title dye were prepared and printed onto Gilbert Bond paper (as NH_4^+ salt) as described in Example 1.

The following results were obtained:

% Dye in ink	OD	Run down (24 h)
2.5	1.113	9

Example 3Preparation of:

Example 1 was repeated except that 2-R acid was used in place of gamma acid in the final coupling reaction.

20

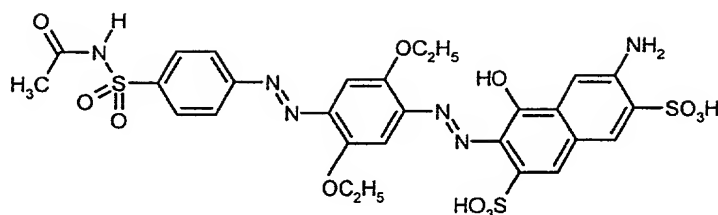
Inks containing the title dye were prepared as in Example 1 except a mixture of glycerol (10%), butyldiglycol (10%), and surfynol 465 (1%) was used in place of N-methyl-2-pyrrolidone and the resultant ink was printed onto Xerox Acid (as NH_4^+ salt) paper using a Epson Color 660 printer.

The following results were obtained:

25

% Dye in ink	OD
6%	1.299

30

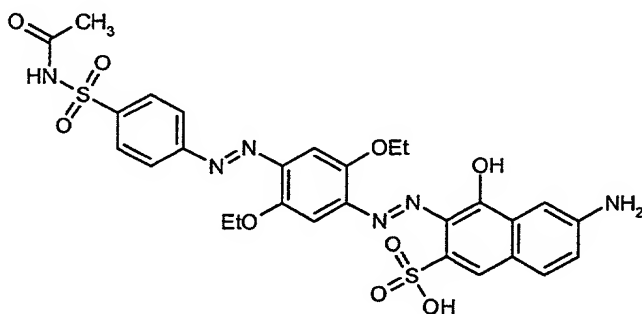
Example 4Preparation of:

Example 1 was repeated except that 2,5-diethoxyaniline was used in place of that 2,5-dimethoxyaniline in the first coupling reaction and 2-R acid was used in place of gamma acid in the final coupling..

Inks containing the title dye were prepared as in Example 1 except a mixture of glycerol (7.5%), thiodiglycol (7.5%), urea (7.5%) and surfynol 465 (1%) was used in place of N-methyl-2-pyrrolidone and the resultant ink was printed onto Xerox Acid (as NH₄⁺ salt) paper using a Canon BJC-2000 printer.

The following results were obtained:

% Dye in ink	OD	Run down (24 h)
3.5	1.075	8.5

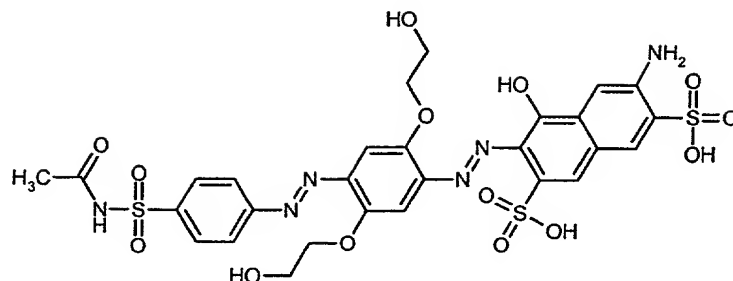
Example 5Preparation of:

Example 1 was repeated except 2,5-diethoxyaniline was used in place of 2,5-dimethoxyaniline in the first coupling reaction.

Inks containing the title dye were prepared and printed onto Xerox Acid paper as described in Example 1.

The following results were obtained:

% Dye in ink	OD	Run down (24 h)
2.5%	1.01	7

Example 6Preparation of:

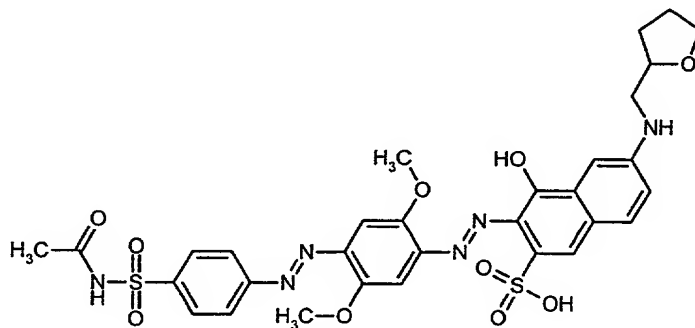
5

Example 1 was repeated except 2,5-di(2-hydroxyethoxy)aniline replaced 2,5-dimethoxyaniline in the first coupling reaction and 2-R acid was used in place of gamma acid in the final coupling.

Inks containing the title dye were prepared as described in Example 5 of PCT/GB00/02280 and printed onto Xerox Acid (as Na⁺ salt) paper as described in Example 1.

The following results were obtained:

% Dye in ink	OD	Run down (24 h)
3	1.07	7.5

Example 7Preparation of:

20

Example 1 was repeated except tetrahydrofurfuryl gamma acid was used in place of gamma acid in the final coupling. Tetrahydrofurfuryl gamma acid was prepared as described in Example 1, stage 2 of WO 9732931 except that tetrahydrofurfurylamine replaced (2-aminoethyl)piperazine.

25

Inks containing the title dye were prepared as described in Example 5 of PCT/GB00/02280 and printed onto Xerox Acid (as Na⁺ salt) paper as described in Example 1.

The following results were obtained:

% Dye in ink	OD	Run down (24 h)
3	1.08	8

Further Inks

The inks described in Tables I and II may be prepared wherein the Dye described in the first column is the Dye made in the above example of the same number. Numbers quoted in the second column onwards refer to the number of parts of the relevant ingredient and all parts are by weight. The inks may be applied to paper by thermal or piezo ink jet printing.

The following abbreviations are used in Table I and II:

PG = propylene glycol

DEG = diethylene glycol

NMP = N-methyl pyrrolidone

DMK = dimethylketone

IPA = isopropanol

MEOH = methanol

2P = 2-pyrrolidone

MIBK = methylisobutyl ketone

P12 = propane-1,2-diol

BDL = butane-2,3-diol

CET = cetyl ammonium bromide

PHO = Na_2HPO_4 and

TBT = tertiary butanol

TDG = thiodiglycol

TABLE I

Example	Dye Content	Water	PG	DEG	NMP	DMK	NaOH	Na Stearate	IPA	MEOH	2P	MIBK
1	2.0	80	5		6	4					5	
2	3.0	90		5	5		0.2					
3	10.0	85	3		3	3				5	1	
4	2.1	91		8								1
5	3.1	86	5					0.2	4			5
6	1.1	81			9		0.5				9	
7	2.5	60	4	15	3	3			6	10	5	4
1	5	65		20					10			
3	2.4	75	5	4		5				6		5
4	4.1	80	3	5	2	10		0.3				
1	3.2	65		5	4	6			5	4	6	5
6	5.1	96								4		
2	10.8	90	5						5			
5	10.0	80	2	6	2	5					4	
1	1.8	80		5					1		15	
1	2.6	84			11						5	
1	3.3	80	2			10				2		6
1	12.0	90				7	0.3		3			
1	5.4	69	2	20	2	1					3	3
1	6.0	91			4						5	

TABLE II

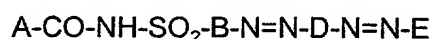
Example	Dye Content	Water	PG	DEG	NMP	CET	TBT	TDG	BDL	PHO	2P	PI2
1	3.0	80	15			0.2					5	
2	9.0	90		5						1.2	5	
3	1.5	85	5	5		0.15	5.0	0.2				
4	2.5	90		6	4					0.12		
5	3.1	82	4	8		0.3					6	
6	0.9	85		10					5	0.2		
7	8.0	90		5	5			0.3				
1	4.0	70		10	4				1		4	11
2	2.2	75	4	10	3				2		6	
3	10.0	91			6						3	
4	9.0	76		9	7		3.0			0.95	5	
5	5.0	78	5	11							6	
7	5.4	86			7						7	
1	2.1	70	5	5	5	0.1	0.2	0.1	5	0.1	5	
1	2.0	90		10								
1	2	88						10				
1	5	78			5			12			5	
6	8	70	2		8			15			5	
1	10	80						8			12	
1	10	80		10								

CLAIMS

1. A compound comprising a disazo chromaphore and an optionally substituted acyl group linked together through a -SO₂-NH- group.

2. A compound according to claim 1 wherein the nitrogen atom of the -SO₂-NH- group is attached to the acyl group through a single covalent bond.

3. A compound according to any one of the preceding claims of Formula (1) and salts thereof:



Formula (1)

wherein:

A is an optionally substituted alkyl or aryl group;

B and D are each independently optionally substituted phenylene or naphthylene;

and

E is optionally substituted naphthylene.

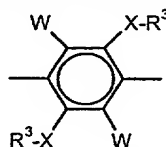
4. A compound according to any one of the preceding claims wherein A is optionally substituted C₁₋₄-alkyl or optionally substituted phenyl.

5. A compound according to any one of claim 3 or claim 4 wherein B is optionally substituted phenylene.

6. A compound according to any one of claims 3 to 5 wherein B is optionally substituted 1,4- phenylene.

7. A compound according to any one of claims 3 to 5 wherein D is optionally substituted 1,4- phenylene.

8. A compound according to any one of claims 3 to 6 wherein D is of the formula:



wherein each:

W independently is H, C₁₋₄-alkyl, alkoxy, polyalkylene oxide, nitro, cyano, amino, sulpho, halo, ureido, SO₂F, PO₄H₂, hydroxy, carboxy, ester, -NR¹R², -COR¹, -CONR¹R² or -SO₂NR¹R², wherein R¹ and R² are each independently H or alkyl;

X independently is O, NH or S;
R³ independently is H or optionally substituted alkyl or optionally substituted aryl.

9. A compound according to any one of claims 3 to 6 wherein D is naphthylene carrying one or more groups selected from hydroxy, amino, sulpho and carboxy.

10. A compound according to any one of claims 3 to 9 wherein E is naphthylene carrying one or more substituents selected from hydroxy, amino, sulpho and carboxy.

11. A compound according to any one of claims 3 to 11 wherein E is a 1-hydroxy-3-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position; or a 1-hydroxy-5-sulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position; or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 8-position or a 1-hydroxy-3,6-disulphonaphth-2-ylene group carrying an optionally substituted amino group at the 7-position

12. A compound according to any one of the preceding claims wherein the pKa of the -NH- group is from 8 to 3.

13. A composition comprising a compound according to any one of claims 1 to 12 and a liquid medium.

14. A composition according to claim 13 which is an ink or liquid dye concentrate.

15. A process for forming an image on a substrate comprising applying an ink according to claim 14 thereto by means of an ink jet printer.

16. A paper, an overhead projector slide or a textile material printed with an ink according to claim 14, with a compound according to any one of claims 1 to 12 or by means of a process according to claim 15.

17. An ink jet printer cartridge comprising a chamber and an ink wherein the ink is in the chamber and the ink is according to claim 14.

18. A method for the colouration of a substrate which comprises treating with a compound according to any one of claims 1 to 12.

ABSTRACT

A compound comprising a disazo chromophore and an optionally substituted acyl group linked together through a -SO₂-NH- group, inks comprising these compounds, ink jet printer cartridges containing these inks and their use in ink jet printing.

5

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RULE 63 (37 C.F.R. 1.63)
DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION IN THE
UNITED STATES PATENT AND TRADEMARK OFFICE

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name and I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the INVENTION ENTITLED:

Disazo dyes and inks containing them

the specification of which

is attached hereto

was filed on as U.S. application serial No.

X was filed as PCT international application No. PCT/GB00/03113 on 14/08/2000
and (if applicable) was amended on

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose all information which is known to me to be material to patentability as defined in 37 C.F.R. 1.56. I hereby claim foreign priority benefits under 35 U.S.C. 119/365 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate filed by me or my assignee disclosing the subject matter claimed in this application and having a filing date (1) before that of the application on which priority is claimed, or (2) if no priority is claimed, before the filing date of this application:

PRIOR FOREIGN APPLICATION(S)

<u>Number</u>	<u>Country</u>	<u>Day/MONTH/Year Filed</u>	<u>Date First Laid Open or published</u>	<u>Date Patented or Granted</u>	<u>Priority claimed Yes No</u>
9921928.9	United Kingdom	16/09/1999			X

I hereby claim the benefit under 35 U.S.C. 120/365 of all United States applications listed below and PCT international applications listed above or below and, if this is a continuation-

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in-part (CIP) application insofar as the subject matter disclosed and claimed in this application is in addition to that disclosed such in the prior applications. I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56 which became available between the filing date of each such prior application and the national or PCT international filing date of this application:

PRIOR U.S. OR PCT APPLICATION(S)Application No. (Serial Code/Serial No.) Day/MONTH/Year FiledStatus(patented, pending abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

And I hereby appoint Pillsbury Winthrop LLP, 1600 Tysons Boulevard, McLean, Virginia 22102 USA, telephone number 861-3000 (to whom all communications should be directed), and the below named persons (of the same address) individually and collectively my attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith and with the resulting patent and I hereby authorize them to act and reply on instructions from and communicate directly with the person/assignee/attorney/firm/organisation who/which first sends/sent this case to them and by who/which I hereby declare that I have consented after full disclosure to be represented unless/until I instruct Pillsbury Winthrop in writing to the contrary.

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